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Plutonium peroxide compound from a New Water-Soluble Peroxo Complex of Plutonium(IV)

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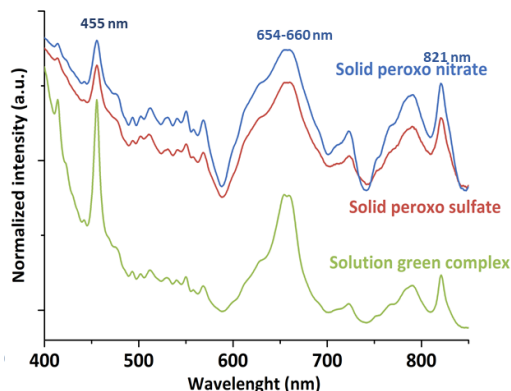
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Over five decades ago the formation of plutonium peroxo complexes were described through the addition of hydrogen peroxide to acidic Pu(IV) solutions leads to the creation of colored complexes, transitioning from brown to red as the concentration of H_2O_2 increases with various proposed structures[1]. Excessive amounts of H_2O_2 relative to Pu(IV) result in the formation of green precipitates, which have been used as precursors for PuO_2 or in waste management processes[2]. Today the only resolved structure for a Pu peroxo compound in the literature involves a dimeric peroxo carbonato compound with bridging μ_2, η_2-O_2 ligands[3]. In this structure, the ten-coordinated Pu atoms are bonded to oxygen atoms from side-on coordinated peroxide ligands and oxygen atoms from bidentate carbonato ligands. In this study we synthesized a new peroxo based tetravalent plutonium complex solution by adding Pu(IV) in highly concentrated H_2O_2 solutions ($n(H_2O_2)/n(Pu)$ ratio > 10) with low Pu(IV) concentrations ($[Pu] < 10$ mM) and a pH of 1-2, a stable green complex solution forms without any precipitate. The resulting solution displayed unique characteristics, with sharp absorption bands at 455 nm and 820 nm and a larger band in the 654-660 nm range (Fig.1). The dried solid results in a green precipitate with an absorption spectra identical to those of the stable green complex in solution. In conclusion, the study highlights the discovery of a soluble green peroxo complex based on Pu(IV), which bears similarities to a green solid compound found during the Manhattan Project. This discovery sheds new light on a chapter of science deeply intertwined with the history of nuclear research.

Figure 1: (a) Vis-NIR absorption spectrum of the green peroxo complex in nitric solution compared to the spectra of the green peroxo compound solid in nitric and sulfate media



[1] R.E. Connick et al., J. Am. Chem. Soc. 71 (1949)

[2] J.A. Leary et al. Ind. Eng. Chem. 51 (1959) 27–31

[3] W. Runde et al. Chem. Commun. (2007) 1728.